

PATENT SPECIFICATION

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COMPLETE SPECIFICATION

Process for the Production of Filled Phenolic Resin Foam Materials

We, DYNAMIT NOBEL AKTIENGESellschaft, of 521 Troisdorf, near Cologne, Germany, a German company, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a process for the production of filled phenolic resin foam materials.

It is known that the properties of synthetic resin compositions can be decisively modified by the addition of inert substances as fillers. Characteristic examples of such modified compositions are the thermosetting moulding compositions, the properties of which depend on the particular fillers used. The usual fillers employed in such compositions, for example, sawdust, ground rock, asbestos powder and short-fibred cellulose, have however proved to be unsuitable for use with phenolic resin foams. Processing difficulties are encountered during the incorporation of such fillers in the foams and also the foamed materials produced have unsatisfactory properties.

It has now been found that any filler substance of relatively large volume and regular or irregular formation, either in the form of an agglomeration of discrete filler particles, the agglomeration including cavities between the particles, or in the form of a loosely bound body of filler, the body containing cavities, can easily be processed with liquid or partially foamed but uncured, foamable phenolic resins to produce foamed materials having properties superior to those of unfilled phenolic resin foam material.

According to the invention there is provided a process for the production of filled phenolic resin foam material, which process comprises wholly or partially filling with

foamable phenolic resin the cavities present between discrete particles of a filler in an agglomeration of such particles or the cavities present in loosely bound bodies formed from such a filler, and thereafter foaming and hardening the phenolic resin *in situ* at a temperature of from 0 to 100°C.

The filler may be, for example, a light inorganic substance, such as vermiculite or expanded clay, inorganic staple fibres or, for example, asbestos or glass, wood shavings, wood wool, waste synthetic resin foam materials or shreds of natural or synthetic fabrics. Depending on the nature and quantity of the filler used, it is possible, for example, to produce filled phenolic foam material having improved mechanical strength values, such as compressive strength, bending strength, tensile strength, impact strength, shearing strength and resistance to abrasion and improved flame resistance. Shaped articles, for example constructional plates, can be formed from the filled phenolic resin foam material.

The entire foaming operation can be carried out with the resins conventionally used for the production of phenolic resin foam and in the temperature range of from 0 to 100°C. in both open and closed moulds. Advantageously the temperature is in the range of from 15 to 60°C.

The phenolic resin used is preferably an alkali condensed resole with a pH value greater than 4, containing a surface-active substance to produce a uniform, cellular structure. Advantageously, the surface active substance is used in a proportion of from 1 to 5 parts by weight per 100 parts of phenolic resin. Preferred surface active agents are polyethylene ethers of monolauric-sorbital ester and condensation products of ethylene oxide and ethyl phenols. A liquid or a solid

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blowing agent may be used to produce the foam and usually an acid-containing hardener is used to cure the resin. The hardener is selected according to the filler actually being used. The foaming of the resin is effected by the transition of the solid or liquid blowing agent into the gaseous state, and the hardening is so controlled that, as soon as the required foam volume is reached, the hardening has progressed to such a degree that there is no collapse of the foam structure.

The following Examples illustrate the invention.

EXAMPLE 1.

1600 g. of a phenolic resin, obtainable commercially under the designation T 612 S, were intimately mixed with 96 ml. of *n*-pentane as blowing agent. (T 612 S is a liquid phenol resol marketed by Dynamit Nobel A.G. and having a solid resin content of 72 to 75%. It is prepared by the condensation of phenol and formalin.) 160 ml. of 70% alcoholic sulphuric acid solution was then added to this mixture as hardener. 5350 g. of expanded clay (particle diameter 10–20 mm.) were wetted with this mixture with stirring and the wetted clay was then introduced with shaking into a rectangular mould (20×30×35 cm.) having an open top. The mould was then tightly closed by means of a cover and heated to 40°C. for 2 hours during which time the foaming and hardening reactions took place.

A body conforming in shape to the mould was thus formed, the compressive strength of the body being increased by 220 to 250% due to the addition of the expanded clay.

The body of foamed phenolic resin material filled with the expanded clay proved to be considerably more resistant to the action of the open flame of a bunsen burner than did a similar body formed from unfilled phenolic resin foam material.

EXAMPLE 2.

2000 g. of the phenolic resin used in Example 1 were intimately mixed with 80 g. of extremely finely powdered calcium carbonate and 100 ml. of *n*-pentane and thereafter with 400 ml. of 20% alcoholic anhydrochloric acid. Immediately after the addition of the hardener, the mixture began to foam due to partial reaction of the hydrochloric acid with the calcium carbonate. 1330 g. of normal commercial wood chips were then wetted with this foam with stirring. The mixture thus obtained was introduced into a mould having a capacity of 25 litres which was then tightly closed with a cover and heated for 2 hours at 40°C. During this heating, the *n*-pentane volatilised causing the mixture to foam further and the resin was cured. By the addition of the wood chips, a moulded body was obtained having a bend-

ing strength higher by about 80% and a tensile strength higher by about 50% than a moulded body produced from an unfilled phenolic resin foam. Also, the impact toughness of the filled body was about 120% higher than that of the unfilled body.

EXAMPLE 3.

A loosely bound body comprising a light wood wool constructional plate, of dimensions 48×23×3 cm. was produced from wood wool (chip width: 3–6 mm., chip length 300–600 mm.), using a synthetic resin binder. The synthetic resin binder was based on a phenolformaldehyde resin. However binders based on ureaformaldehyde resins, polyester resins or epoxide resins may be used. The synthetic resin binder was such as to cause a bonding of the individual wood wool fibres at their mutual points of contact leaving cavities between the individual fibres. 241 g. of a foamable mixture consisting of 200 g. of phenolic resin obtainable commercially under the designation T 612 S, 14 ml. of monofluorotrichloromethane as blowing agent and 18 ml. of 20% alcoholic hydrochloric acid as hardener were introduced into a shallow, upwardly open box-type mould, the base area of which was 50×25 cm. The thickness of the bed of mixture in the horizontally positioned mould was about 2 mm. The constructional plate was then immediately placed in the mixture and fixed in position by means of a cover fitted to the mould. The mould and its contents were then placed for 60 minutes in a heated cupboard at about 35°C. During this period, the foam penetrated into those cavities of the plate which were accessible to it and was cured. The product thus obtained had a heat conductivity of $\lambda=0.04$ Kcal/mh°C. at 20°C. The heat conductivity λ is the amount of heat conducted per unit time across a temperature gradient of 1°C. (1 Kcal./m.h.°C. is equivalent to 0.672 BTU/ft.h.°F.) A similar light wood wool constructional plate (weight per unit volume about 430 kg/m³) in which the cavities were not filled with foam material had a heat conductivity of $\lambda=0.07$ Kcal/mh°C. at 20°C.

WHAT WE CLAIM IS:—

1. Process for the production of filled phenolic resin foam material, which process comprises wholly or partially filled with foamable phenolic resin the cavities present between discrete particles of a filler in an agglomeration of such particles or the cavities present in loosely bound bodies formed from such a filler, and thereafter foaming and hardening the phenolic resin *in situ* at a temperature of from 0 to 100°C.

2. Process according to Claim 1, wherein the foamable phenolic resin is partially pre-foamed before being used to fill or partially fill the cavities.

3. Process according to Claim 1 or 2, wherein the foamable phenolic resin comprises an alkali condensed resole of pH greater than 4 and a surface active agent.
- 5 4. Process according to Claim 3, wherein the surface active agent is present in an amount of from 1 to 5 parts by weight per 100 parts by weight of phenolic resin.
- 10 5. Process according to any preceding claim, wherein the foam is produced using n-pentane as blowing agent.
6. Process according to any one of Claims 1 to 4, wherein the foam is produced using monofluorotrichloromethane as blowing agent.
- 15 7. Process according to any preceding claim, wherein the filler is a light inorganic substance.
8. Process according to Claim 7, wherein the filler is vermiculite.
- 20 9. Process according to Claim 7, wherein the filler is expanded clay.
10. Process according to any one of Claims 1 to 6, wherein the filler comprises inorganic staple fibres.
- 25 11. Process according to Claim 10, wherein the fibres are asbestos fibres.
12. Process according to Claim 10, wherein the fibres are glass fibres.
13. Process according to any one of Claims 1 to 6, wherein the filler is wood shavings or wood wool. 30
14. Process according to any one of Claims 1 to 6, wherein the filler is waste synthetic resin foam.
15. Process according to any one of Claims 1 to 6, wherein the filler is shredded natural or synthetic fabric. 35
16. Process according to Claim 1, substantially as described in any one of the Examples. 40
17. A filled phenolic resin foam material whenever produced by the process claimed in any preceding claim.
18. A shaped article whenever prepared from the filled phenolic resin foam material claimed in Claim 17. 45

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